

Pulsed-gate measurements of the singlet-triplet relaxation time in a two-electron double quantum dot

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A pulsed-gate technique with charge sensing is used to measure the singlet-triplet relaxation time for nearly-degenerate spin states in a two-electron double quantum dot. Transitions from the (1,1) charge occupancy state to the (0,2) state, measured as a function of pulse cycle duration and magnetic field, allow the (1,1) singlet-triplet relaxation time ($\geq 70 \mu\text{s}$) and the (0,2) singlet-triplet splitting to be measured. The use of charge sensing rather than current measurement allows long relaxation times to be readily probed.

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Semiconductor quantum dots are promising systems for the manipulation of electron spin because of the relative ease of confining and measuring single electrons [1]. In order to make use of the spin degree of freedom as a holder of either classical or quantum information, it is first necessary to understand and characterize the mechanisms that lead to spin relaxation and decoherence.

Previous studies of spin relaxation in quantum dots have focused on systems with large energy splittings of the relevant spin states, either singlet and triplet two-electron states, split by $\sim 600 \mu\text{eV}$ [2], or Zeeman states, split by $\geq 200 \mu\text{eV}$ [3, 4, 5]. In these cases, spin-orbit mediated processes are expected to dominate spin relaxation [6, 7], while processes involving hyperfine coupling of electron spin to nuclei are relatively ineffective for splittings greater than the hyperfine coupling scale $\sim 0.1 \text{ meV} / \sqrt{N_E} \sim 1 \mu\text{eV}$, where N_E is the effective number of nuclei interacting with the electron spin [8, 9, 10]. Spin relaxation between nearly degenerate electronic states is particularly relevant to the problem of controlled entanglement, as singlet-triplet splitting goes to zero as the entangled spins become spatially separated.

In this Letter, we present pulsed-gate measurements of the singlet-triplet relaxation times and splittings in a two-electron double quantum dot. Spin states of the weakly-coupled (1,1) charge configuration (integer pairs specify the equilibrium charge occupancy on the left and right dot) are nearly degenerate. A spin selection rule impedes the transition from (1,1) spin triplets to the (0,2) singlet. To probe spin relaxation, the double dot is initialized, then loaded into the (1,1) state, then placed in a configuration where (0,2) is the preferred charge distribution in equilibrium. This transition probability is measured from a calibrated charge sensing signal as a function of the period of this three-step cycle. The resulting characteristic time of $\sim 70 \mu\text{s}$ sets the time scale for of the spin-blockaded relaxation channels from (1,1) to the singlet state of (0,2). Measurements of the (1,1) to (0,2) transition probability as a function of detuning, ϵ , show a strong dependence on perpendicular magnetic field, B_\perp ,

reflecting the singlet-triplet splitting of the (0,2) charge state, J as discussed below.

Samples are fabricated from a GaAs/Al_{0.3}Ga_{0.7}As heterostructure grown by molecular beam epitaxy. Electron beam lithography and liftoff are used to create Ti/Au gates that deplete a 100 nm deep two-dimensional electron gas with electron density $2 \times 10^{11} \text{ cm}^{-2}$ and mobility $2 \times 10^5 \text{ cm}^2/\text{V}\cdot\text{s}$. Gates 2–6 and 12 form the double quantum dot [see Fig. 1(a)]. Gates 2 and 6 are connected via bias tees to dc voltage sources and to pulse generators through coax cables with $\sim 20 \text{ dB}$ of inline attenuation [11]. A QPC charge detector is created by depleting gate 1. Gates 7–11 are unused. The QPC conductance, G_S , is measured using standard lock-in amplifier techniques

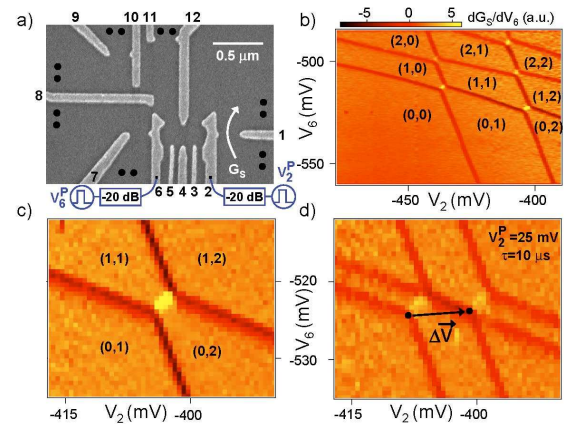


FIG. 1: (a) SEM image of a device identical in design to the one used in this experiment. Gates 2–6 and 12 define the double dot. • denotes an ohmic contact. (b) Large-scale plot of dG_S/dV_6 as a function of V_2 and V_6 . Charge states are labelled (M, N) , where $M(N)$ is the time averaged number of electrons on the left (right) dot. (c) Zoom in near the (1,1) to (0,2) charge transition. (d) 25 mV pulses with a 50% duty cycle and 10 μs period are applied to the coax connected to gate 2. This results in two copies of the charge stability diagram shifted relative to one another by ΔV .

with a 1 nA current bias at 93 Hz. The electron temperature, $T_e \sim 135$ mK, was determined from Coulomb blockade peak widths. We present results from a single sample; a second sample with slightly larger lithographic dimensions gave qualitatively similar results [12].

QPC charge sensing is used to determine the absolute number of electrons in the double dot. Figure 1(b) shows a large-scale charge stability diagram for the double dot. As electrons enter or leave the double dot, or transfer from one dot to the other, G_S changes, resulting in sharp features in dG_S/dV_6 (numerically differentiated) [12, 13]. In the lower left corner of Fig. 1(b), the double dot is completely empty. As the gate voltages are made more positive, electrons are added to the double dot. We will focus on the two-electron regime near the (1,1) to (0,2) charge transition [Fig. 1(c)]. In this location, charge transport in a double dot shows a striking asymmetry in bias voltage due to spin selection rules (Pauli blocking) [14, 15]. At forward bias, transitions from the (0,2)_S to the (1,1) singlet state (1,1)_S are allowed. However, for reverse bias, the (1,1) to (0,2) transition can be blocked if the (1,1) state forms a (1,1)_T because the (0,2)_T state resides outside the transport window due to the large singlet-triplet splitting in (0,2). This asymmetry results in current rectification, which is used in present pulsed gate measurements.

The double dot is electrostatically driven by applying pulses to gates 2 and 6. Figure 1(d) shows a charge stability diagram acquired with square pulses applied to gate 2 ($V_2^P = 25$ mV, 50% duty cycle, period $\tau = 10$ μ s). This results in two copies of the charge stability diagram, the right-most (left-most) charge stability diagram reflects the ground state charge configuration during the low (high) stage of the pulse sequence. The gate-voltage offset between the charge stability diagrams, $\Delta \vec{V}$, is used to calibrate pulse amplitudes. Additional calibrations are performed for gate 6, which primarily shifts the honeycomb in the vertical direction (not shown). A linear combination of pulses on gates 2 and 6 can be used to shift the stability diagram in any direction in gate space.

As a control, we compare a forward pulse sequence, where spin selection rules are expected to be important, with a reversed pulse sequence that does not involve spin selective transitions. The forward pulse sequence (Fig. 2(a)) begins with the gates at point E for 10% of the period, emptying the second electron from the double dot, leaving the (0,1) charge state. The gates then shift to the reset point R for the next 10% of the period, which initializes the system into the (1,1) configuration. The interdot tunnel coupling, t , is tuned with $t \leq k_B T$ so that the (1,1) singlet-triplet splitting $j \sim 4t^2/U \ll k_B T$, where U is the single dot charging energy. Due to this degeneracy, and at low fields such that $|g|\mu_B B < k_B T$ ($|g| \sim 0.44$ for GaAs), we expect to load into (1,1)_S or any of the three (1,1)_T states with equal probability. For the final 80% of the period, the gates are at the measurement point M where (0,2)_S is the ground state. Figure 2(b) illustrates the possible (1,1) to (0,2) transitions. If the R

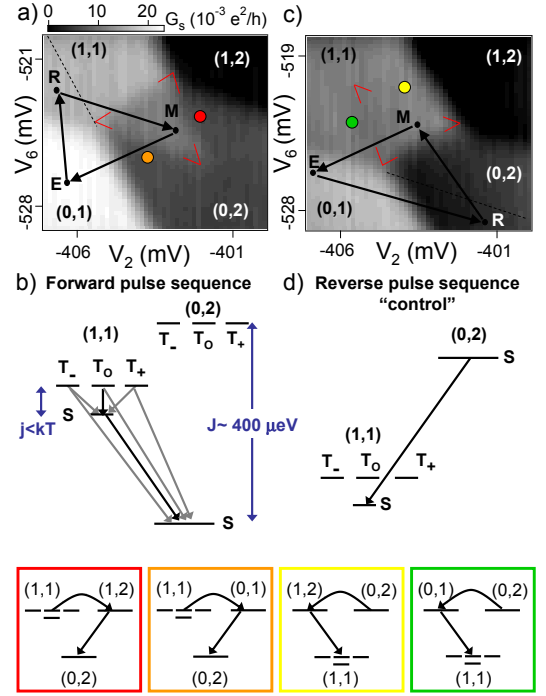


FIG. 2: (a) Sensor conductance, G_S , as a function of V_2 and V_6 while applying the forward pulse sequence (see text) with $\tau = 10$ μ s and $B_\perp = 100$ mT. Spin-blocked transitions result in some (1,1) charge signal in the (0,2) pulse triangle (bounded by the red lines). Outside of the pulse triangle it is possible to access other charge states. This relaxes the spin blockade (see color-coded level diagrams). (b) Level diagram illustrating the possible transitions from (1,1) to (0,2). Black arrows indicate fast transitions, grey arrows indicate spin blocked transitions. (c) G_S as a function of V_2 and V_6 while applying the reverse “control” pulse sequence with $\tau = 10$ μ s and $B_\perp = 100$ mT. The (0,2)_S to (1,1)_S transition is not spin blocked. As a result, there is no detectable pulse signal in the pulse triangle (bounded by the red lines). (d) Level diagram illustrating the (0,2) to (1,1) transition. A best-fit plane has been subtracted from the data in (a),(c) to remove signal from direct gate to QPC coupling.

step loads the (1,1)_S state, tunneling to (0,2)_S occurs on a timescale given by the interdot tunneling rate, $\Gamma(\epsilon)$ (We estimate the slowest $\Gamma(\epsilon)(1 \mu s)^{-1}$ from finite bias data [15]). If the $m_s = 0$ (1,1) triplet state (1,1)_{T0} is loaded, it dephases into (1,1)_S on a timescale of T_2 (expected to be ≤ 100 ns [10, 16, 17]) followed by a direct transition to (0,2)_S. About half the time the R step will load the $m_s = 1$ (1,1) triplet state (1,1)_{T+} or the $m_s = -1$ (1,1) triplet state (1,1)_{T-}. At low B_\perp , (0,2)_T is inaccessible, and a transition from (1,1)_{T+} or (1,1)_{T-} to (0,2) requires a spin flip and will be blocked for times shorter than the singlet-triplet relaxation time τ_{ST} .

In Fig. 2(a) the average charge sensor signal, G_S , is measured as a function of the dc gate voltages V_2 and V_6 , while a pulse sequence is repeated. This has the effect of translating the points E, R, and M throughout the charge stability diagram, keeping their relative positions

constant. Because most of the time is spent at point M, the grayscale data primarily map out the ground state population for this point, with plateaus at $G_S \sim 0.0, 6.0, 16,$ and $23 \times 10^{-3} e^2/h$ indicating full population of the (1,2), (0,2), (1,1), and (0,1) charge states respectively. The pulse data differs from ground state data only when point M resides in the triangle defined by the (1,1) to (0,2) ground state transition and the extensions of the (1,1) to (0,1) and (1,1) to (1,2) ground state transitions (bounded by the red marks in Fig. 2(a)). Within this “pulse triangle” transitions from (1,1) to (0,2) may be blocked as described above, and the charge sensor registers a conductance intermediate between the (1,1) and (0,2) plateaus. If M moves above the pulse triangle (red dot in Fig. 2(a)), the (1,1) to (0,2) transition can occur sequentially via (1,2) with no interdot tunneling: a new electron enters the right dot, then the electron in the left dot leaves. Likewise, if M moves below the pulse triangle (orange dot in Fig. 2(a)) the transition can occur via (0,1): the left-dot electron leaves, then a new electron enters the right dot. By similar logic, point R must be to the left of the (0,1) to (0,2) transition extension (dotted line in Fig. 2(a)) to avoid resetting through (0,2) and preferentially loading (1,1)_S. Figure 2(a) shows a signal of $11 \times 10^{-3} e^2/h$ in the pulse triangle for $\tau = 10 \mu s$, which indicates that approximately 50% of the time the dots remain in (1,1) even though (0,2) is the ground state. This is direct evidence of spin-blocked (1,1) to (0,2) transitions.

If the the pulse sequence is reversed, so that the reset position R occurs in (0,2), where only the singlet state is accessible, and M occurs in (1,1), tunneling from R to M should always proceed on a time scale set by the interdot tunnel coupling, since the (0,2)_S to (1,1)_S transition is not spin blocked. As anticipated, no signal is seen in the pulse triangle for this reversed “control” sequence (Fig. 2(c)).

Spin selectivity of the forward pulse sequence in Fig. 3 can be used to measure J as a function of B_\perp [18]. Figure 3(a) shows G_S as a function of V_2 and V_6 while applying the forward pulse sequence with $B_\perp = 1.2$ T and $\tau = 10 \mu s$. For these data, (0,2)_T resides outside of the pulse triangle (JE_M , the mutual charging energy) and the (1,1)_T to (0,2) transitions are spin blocked. For $B_\perp = 1.4$ T [Fig. 3(b)] the (0,2)_T state is low enough in energy that the (1,1)_T states can directly tunnel to the (0,2)_T manifold at high detunings. Now (1,1) to (0,2) tunneling can proceed, and there is no longer a (1,1) charge signal in the (0,2) region of the pulse triangle at high detuning. This cuts off the tip of the pulse triangle. The spin-blocked region continues to shrink as B_\perp is increased. From these data, we find $J \sim 340, 280,$ and $180 \mu eV$ for $B_\perp = 1.4, 1.6,$ and 1.8 T, respectively [19].

The time dependence of the charge sensing signal can be investigated by varying τ , the overall period of the cycle. Figure 4(a) shows G_S as a function of V_2 and V_6 acquired using the forward pulse sequence with $\tau = 8 \mu s$ at $B_\perp = 100$ mT. A clear pulse signal is observed in

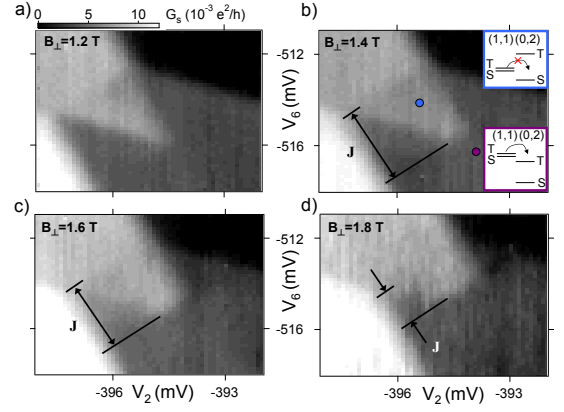


FIG. 3: Forward pulse sequence results with $\tau = 10 \mu s$. (a–d) G_S as a function of V_2 and V_6 for increasing B_\perp . B_\perp reduces J . Eventually (0,2)_T is lowered into the “M” pulse window [inset of (b)]. At this point, (1,1)_T to (0,2)_T transitions are energetically possible and the transition from the (1,1) to (0,2) charge state is no longer “spin blocked”. This cuts off the tip of the pulse triangle in (0,2), see (b). A best-fit plane has been subtracted from the data in (a)–(d).

the pulse triangle. As τ is increased, the pulse signal decreases as shown in (b–c). G_S is measured inside the pulse triangle (V_2, V_6 held fixed at $-403, -523.8$ mV, respectively) and is plotted as a function of τ in Fig. 4(d). In (1,1), $G_S \sim 20 \times 10^{-3} e^2/h$, whereas outside the pulse triangle in (0,2), $G_S \sim 10 \times 10^{-3} e^2/h$. For small τ , $G_S \sim 15 \times 10^{-3} e^2/h$ in the pulse triangle. At long τ , G_S approaches $10 \times 10^{-3} e^2/h$ in the pulse triangle, which indicates complete transfer from the (1,1) to (0,2) charge state.

These data are consistent with spin-blocked transitions preventing the (1,1) to (0,2) charge transition. Approximately 50% of the time, the (1,1) R pulse loads into either (1,1)_{T+} or (1,1)_{T-}. These states may relax into the (1,1)_S state and then tunnel to (0,2)_S on a timescale set by τ_{ST} . For $\tau \ll \tau_{ST}$, we expect that 50% of the (1,1) to (0,2) transitions will be spin-blocked, resulting in $\sim 50\%$ (1,1) pulse signal in the (0,2) pulse triangle. For $\tau \gg \tau_{ST}$, the (1,1)_{T+} and (1,1)_{T-} states have ample time to relax to (1,1)_S, after which a (1,1)_S to (0,2)_S transition can take place. Thus, for τ long compared to τ_{ST} , the pulse signal to approaches the (0,2) level, indicating full transfer from (1,1) to (0,2). In the intermediate τ regime, the sensor signal due to spin-blocked transitions decays as a function of time on a timescale that is characteristic of τ_{ST} .

The experimental data in Fig. 4(d) are fit assuming exponential singlet-triplet relaxation. Due to the slow measurement rate of the charge sensor (~ 100 ms), G_S is proportional to the time-averaged occupation of the left dot. Modeling an exponential decay of the sensing signal weighted over the 80% of the cycle corresponding to the (0,2) measurement gives $G_S(\tau) = A + B(\tau_{ST}/\tau)(1 - e^{-0.8\tau/\tau_{ST}})$, where A is the conductance asymptote at

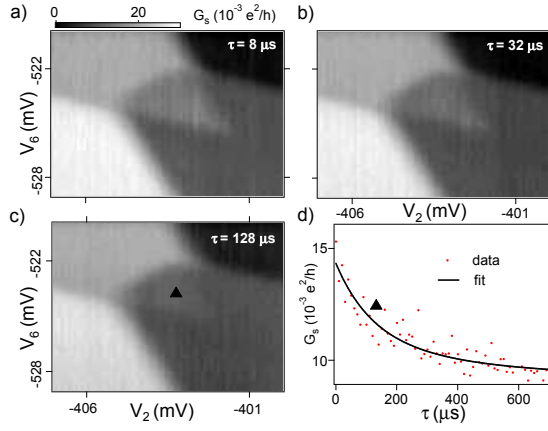


FIG. 4: Forward pulse sequence results with $B_{\perp}=100$ mT. (a)–(c) G_S as a function of V_2 and V_6 for increasing pulse periods, τ . For longer periods, singlet-triplet relaxation occurs and the (1,1) to (0,2) transition proceeds, reducing the signal in the (0,2) pulse triangle. (d) G_S with $V_2=-403.0$ mV, $V_6=-523.8$ mV (gate voltage position indicated by the black triangle in (c)) measured as a function of τ . $G_S=10 \times 10^{-3} e^2/h$ in the (0,2) charge state, while $G_S=20 \times 10^{-3} e^2/h$ in the (1,1) charge state. For short τ , the (1,1) to (0,2) transition is blocked approximately half of the time, resulting in a pulse signal of $15 \times 10^{-3} e^2/h$ in the (0,2) pulse triangle. The (1,1) to (0,2) transition probability increases (sensor signal decreases) with τ due to spin-relaxation with a characteristic time scale of $70 \pm 10 \mu s$. A best-fit plane has been subtracted from the data in (a)–(c).

long times (full occupation of the (0,2) state) and B is the additional conductance in a short pulse due to the blocked states (approximately 50% of the (0,2) to (1,1) step height). The best fit to the data in Fig. 4(d) gives $A=0.009 e^2/h$ and $B=0.007 e^2/h$, consistent with these expectations. In the center of the pulse triangle the best-fit τ_{ST} reaches a maximum of $\tau_{ST}=70 \pm 10 \mu s$. Near the

(1,1) to (0,2) transition, τ_{ST} decreases, to $20 \pm 5 \mu s$ at $V_2=-403.8$ mV and $V_6=-523.0$ mV. Closer to the tip of the pulse triangle, τ_{ST} decreases due to thermally activated exchange with the leads (See Fig. 2, red and orange diagrams), thus the $70 \mu s$ relaxation time within the (1,1) manifold [20].

In GaAs quantum dots, spin-orbit and hyperfine interactions are expected to be the dominant mechanisms for spin relaxation. The spin-orbit contribution to T_1 depends strongly on B and theory predicts $T_1=10$ ms for $B=1$ T [7]. The hyperfine spin relaxation rate is $\Gamma_{HF}=\Gamma_{ph}\left(\frac{E_n}{j}\right)2\frac{\gamma_n G_C}{(2T)^2 N_E}$ where $\Gamma_{ph} \approx 4 \times 10^7 s^{-1}$ reflects the electron-phonon coupling, $E_n=0.135$ meV is the nuclear energy for a fully polarized system, $G_C=1.25$ is derived from the correlation of the nuclear spins, $\gamma_n=0.045$ depends on wavefunction overlap, the nuclear spin $I=3/2$, and $N_E \sim 10^4$ [9]. Our sample is tuned with $j \sim 4t/U \ll k_B T$. In this case, the Zeeman splitting between the $m_s=0$ state and $m_s=\pm 1$ states sets the relevant energy scale. Using $j \sim 2.5 \mu eV$ gives $1/\Gamma_{HF}=15 \mu s$, roughly consistent with the experimental results. The relaxation times found here can be compared with those obtained by Fujisawa *et al.* who measured $\tau_{ST}=200 \mu s$ in a vertical dot [2]. In those measurements, the sizable singlet-triplet splitting implies that spin-orbit processes rather than nuclear processes dominant, in contrast to the present experiments.

Acknowledgments

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 - [19] The conversion from gate voltage to energy is determined by measuring finite bias triangles.
 - [20] $\Gamma(\epsilon)$ is set to be much faster than the characteristic relaxation times that we measure in this experiment. $\Gamma(\epsilon)$ varies strongly with ϵ (see ref. 13). The fact that τ_{ST} only varies by roughly a factor of three in the pulse triangle also indicates that the (1,1)-(0,2) transitions are spin-blocked and not limited by a slow $\Gamma(\epsilon)$.